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PHOTOCHEMICAL METHODS FOR PURIFYING WATER

C. Y. Cha, et al

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16. Abstract

The kinetics of photochemical oxidation of organic carbon pollutants, with and without sensitization by chlorine, have been determined. The results, which are suitable for use in designing reactors for any type of lamp and lamp-reactor geometry, are based upon data obtained for secondary effluents from Sacramento area municipal treatment plants.

A trough reactor with parabolic reflector was ~~found to be~~ the most suitable design for a large-scale, tertiary treatment plant based upon photochemical oxidation. Cost calculations were made for reducing the TOC from 10 to 4 mg/liter and maintaining a chlorine concentration of about 0.8 mg/liter. For a capacity of one-million gallons per day of secondary effluent, treatment costs were 89¢ per 1000 gallons with a total plant investment of $563,000. (EPA abstract)

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PHOTOCHEMICAL METHODS FOR PURIFYING WATER

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Project 17020 EVQ

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ABSTRACT

(including Conclusions and Recommendations)

The kinetics of photochemical pollutant removal and chlorine disappearance have been developed in a form suitable for use for various lamps, reactor-lamp geometries, and wastewaters. Numerical kinetic equations also are presented for secondary effluents from the Sacramento, California area. The rate of pollutant removal is directly proportional to the UV energy absorbed by pollutant. The rate of disappearance of free available chlorine is directly proportional to the UV energy absorbed, not only by free available chlorine but by pollutant. The rate of pollutant removal for a conventional, low-pressure mercury lamp is about 6-fold larger than the rate for a high-pressure mercury lamp, for the same light absorption.

A trough reactor with parabolic reflector was found to be most suitable for tertiary treatment. Design equations for electric power and chlorine requirements have been developed. Also an equation for calculating the fraction of UV energy which reaches the surface of water has been obtained. Low- and high-pressure lamps were compared, and the G64T6 low-pressure mercury lamp (General Electric Co.) was chosen as the most effective UV-energy source now available. Optimum values for chlorine concentration and reactor-reflector geometry have been determined for tertiary treatment.

Costs have been calculated approximately for a plant (capacity of one-million gallons per day of secondary effluent) to reduce TOC from 10 to 4 mg/liter. Total plant investment was estimated to be $563,200. Treatment cost was estimated at $323,600 per year, or 89.5¢/(1,000 gal.), of which the major costs were; labor, 17.8¢/(1,000 gal.); electricity, 13.9¢/(1,000 gal.); lamp replacement, 34.2¢/(1,000 gal.). The treatment cost could be reduced significantly by improving the intensity, lifetime, and UV output per unit power input of lamp.

The treatment cost of 89.5¢/(1,000 gal.) is much greater than the 11.8¢/(1,000 gal.) estimated in a previous investigation (1), mainly due to differences in quantum yield.

Research and development data in detail are presented in four articles (2-5), which have been published in widely distributed technical journals. The body of the report contains design calculations and results based upon the research data

It should be emphasized that the numerical results presented here apply only for secondary effluent from Sacramento area treatment plants. It is highly desirable to apply the concepts developed here to a number of other secondary effluents of different properties. Also, for improvement and better understanding of the photochemical process it is

important to determine specific kinetic equations for removal of various types of organics (i.e. acids, phenols, ketones, etc.). With this information it should be possible to design the optimum photochemical treatment for a given wastewater.

A second area where more research is needed, prior to careful economic evaluation of photochemical processing, is small pilot-plant studies of reactor-lamp-reflector geometry. This work is necessary in order to substantiate efficiency calculations, such as those considered in Section IV of this report.

This report was submitted in fulfillment of Project Number 17020 EVQ, under the partial sponsorship of the Environmental Protection Agency.

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II. INTRODUCTION

The primary objective of this work was to develop and evaluate the economics of a continuous photo-oxidation process for reducing the organic pollutants in secondary effluents from municipal wastewater treatment plants. To determine the technical feasibility of employing the photo-oxidation process for tertiary treatment, it is first necessary to develop the reaction kinetics of pollutant removal. Once rate equations are obtained, reactor design calculations are required for process evaluation. Since the cost of the photo-oxidation process can depend heavily upon the efficiency of energy utilization, it is necessary to analyze the reflector-lamp-reactor geometry before economic evaluation.

This report consists of three major parts: the first describes the reaction kinetics of organic pollutant removal [the details of this part are given in references (2), (3), (4)]; the second part discusses process design for tertiary treatment; the third part is an economic evaluation and utilizes the results published in reference (5).

III. KINETICS OF ORGANIC POLLUTANT REMOVAL

Rate of Pollutant Removal Due to $A_p I_{tot}$ and C_0; Unsensitized Rate

The rate of pollutant disappearance was found to be first order in the amount of radiation absorbed, directly proportional to oxygen concentration at low C_0, and independent of C_0 at high oxygen concentrations. Reference (2) describes details of procedure to determine the rate, using a high-pressure mercury lamp. The final form of the rate equation obtained for a high-pressure lamp is

$$\left(\Omega_p\right)_{Hp} = \frac{1.47 \times 10^{-3}\, C_0}{6.5 \times 10^{-8} + C_0} A_p I_{tot} \qquad (1)$$

When the shorter wave length radiation of a low-pressure mercury lamp is used, the quantum yield increases 14 times. The experimental data are given in Reference (4). The final rate equation obtained when using a low-pressure lamp is

$$\left(\Omega_p\right)_{Lp} = \frac{2.07 \times 10^{-2}\, C_0}{6.5 \times 10^{-8} + C_0} A_p I_{tot} \qquad (2)$$

It will be seen later that the rate of pollutant disappearance increases significantly when chlorine is applied.

Rate of Pollutant Removal Due to $A_p I_{tot}$ and C_{Cl}; Sensitized Rate

When chlorine is added to secondary effluent, the rate of pollutant disappearance increases significantly for both high- and low-pressure lamps. The rate, Ω_p, consists of two contributions: one for the non-sensitized reaction with oxygen, given by Eqs. (1) and (2), and a second for reaction between pollutant and available chlorine. The experimental data and method of correlation for the rate, using the high-pressure mercury lamp, are given in detail in (2) and (3). The final expression for the sensitized rate for the high-pressure lamp is

$$\left(\Omega_p\right)_{Hp} = \left[\frac{1.47 \times 10^{-3}\, C_0}{6.5 \times 10^{-8} + C_0} + \frac{2.1 \times 10^{-2}\, C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}}\right] A_p I_{tot} \qquad (3)$$

The sensitized rate also can be improved by using the low-pressure mercury lamp. The experimental data for a low-pressure lamp are given

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in (4). The rate equation for the low-pressure lamp is

$$\left(\Omega_p\right)_{Lp} = \left[\frac{2.07 \times 10^{-2}\, C_O}{6.5 \times 10^{-8} + C_O} + \frac{1.01 \times 10^{-1}\, C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}}\right] A_p I_{tot} \qquad (4)$$

Comparing Eq. (3) with (4), we see that the quantum yield is approximately six times greater for the low-pressure mercury lamp.

The rate of free available chlorine disappearance was found to be directly proportional to the UV energy absorbed by both free available chlorine and pollutant. The method of obtaining the rate, Ω_{Cl}, for low- and high-pressure lamps is described in (4). The final results are as follows:

for a high-pressure lamp

$$\left(\Omega_{Cl}\right)_{Hp} = 0.37\, A_{Cl} I_{tot} + \frac{0.062\, C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} A_p I_{tot} \qquad (5)$$

for a low-pressure lamp

$$\left(\Omega_{Cl}\right)_{Lp} = 0.52\, A_{Cl} I_{tot} + \frac{0.057\, C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} A_p I_{tot} \qquad (6)$$

Comparison of Eq. (5) with (6) shows that the low-pressure lamp gives about 1.4-times higher rate, for the same intensity of radiation.

In order to avoid the complication of the effects of specific components of secondary effluent on kinetics, the rate equations were formulated in terms of A_p. However, it would be desirable in future work to study a variety of secondary effluents in order to accumulate information on the relation between A_p and measurable properties of the effluents. Nevertheless, A_p can be calculated from transmittance measurements and thus not require such a relation. For design calculations for a specific secondary effluent, A_p should be expressed in terms of concentration of pollutants. A_p depends on the composition of secondary effluent and on the amount of chlorine added initially. This dependence for a Sacramento effluent will be discussed in the following section.

Note that the rate equations given in this part are not restricted to any particular reactor-reflector geometry.

IV. PROCESS DESIGN

Reactor Type and Design Conditions

Since methods for obtaining the rate of removal of organic carbon [Eqs. (3) and (4)] and for the efficiency of UV-energy transfer to the surface of water (5) have been obtained, it is now possible to design a photoreactor for a tertiary treatment process. First, it is necessary to decide upon a type of reactor which is feasible for photochemical treatment on a large scale. The secondary effluent is a colloidal solution which may deposit on walls. These deposits will decrease the transmittance of light so that the fraction of light energy which reaches the surface of the reacting solution will be reduced. Therefore, a system having no surfaces between the lamp and reactor is desirable. Such a system is also attractive from the economic point of view since it would be expensive to construct transparent walls for a large-scale system.

An attractive type is the trough reactor (Figure 1) which is irradiated from a lamp above the water and surrounded by a parabolic reflector at the top. The system may operate as a stirred-tank reactor or as an integral-flow reactor. The integral arrangement would be more suitable for tertiary treatment. This follows from the following reasoning. Since the electric-energy requirement is likely to be a major cost in photoreactors, it is more economical to arrange the reactor such that a constant, large fraction of light energy is absorbed along the flow path in the reactor. This may be achieved by continuously increasing the depth as pollutant concentration decreases due to oxidation. This can be accomplished in the integral-flow type, but not in a stirred-tank reactor. At low concentrations of free available chlorine the rate increases directly with C_{Cl}. Therefore, multiple injections of small quantities of chlorine along the flow path, always operating at low C_{Cl}, are desirable. The contribution of oxygen to the rate is insignificant for a high-pressure lamp but not negligible for low-pressure lamp operation. Hence, multiple injections of oxygen are also desirable. However, it would be more economical to use air instead of oxygen, considering that the effect of concentration of oxygen on the rate is much smaller than that of chlorine. Therefore, the following design calculations will be based upon an integral-flow, trough reactor with multiple injections of chlorine and air.

The basis for design calculations are:

1. The concentrations of chlorine and oxygen are maintained reasonably constant throughout the reactor by supplying chlorine and air along the reactor length. The numerical value for oxygen concentration is 0.28×10^{-6} g moles/cm^3, corresponding to saturation of water with air at room temperature.

2. A constant fraction of UV energy is absorbed along the reactor flow length (length of trough) by changing the depth of the reactor. The

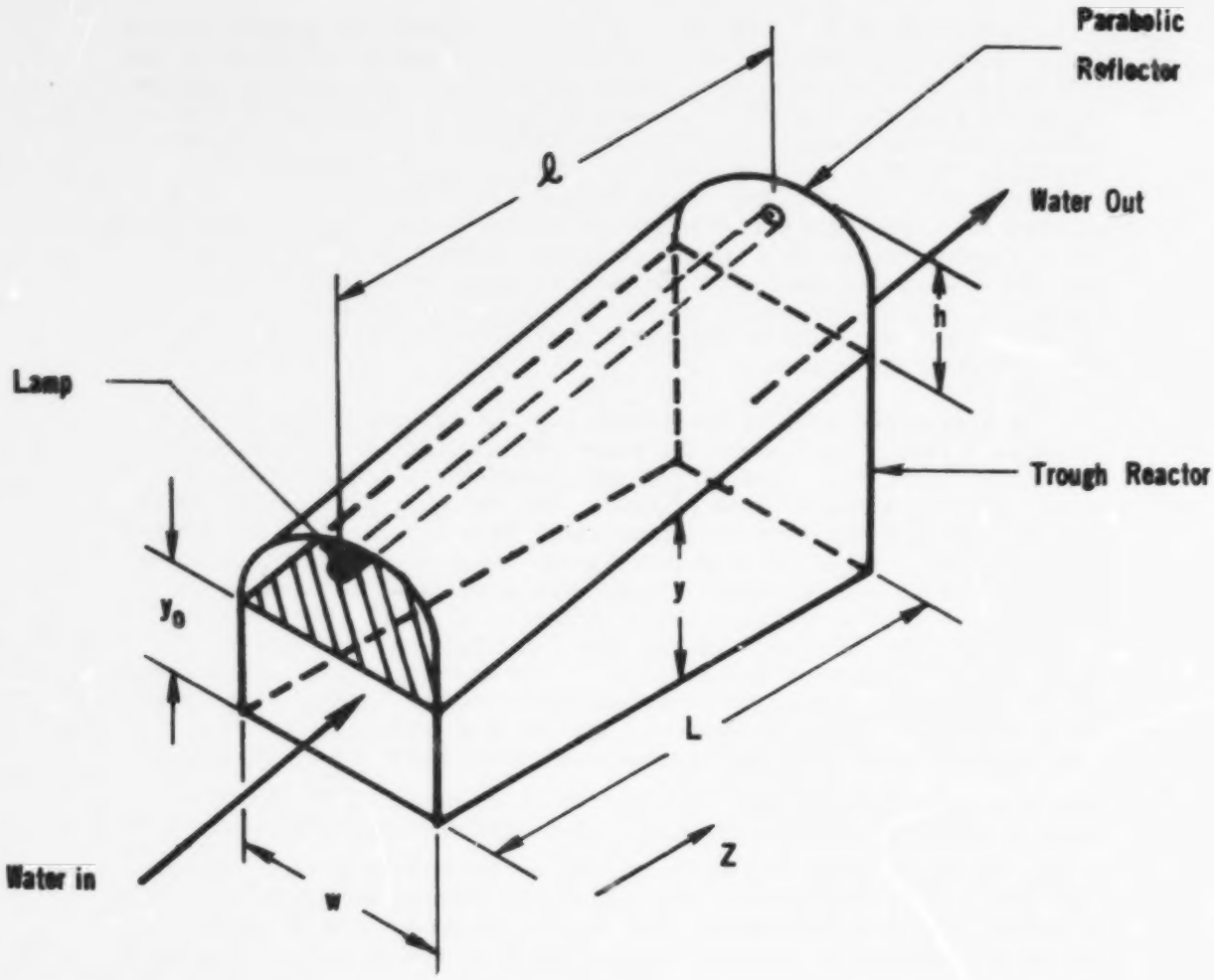


Figure 1. Trough Reactor with Parabolic Reflector

numerical values for chlorine concentration and fraction of UV energy will be determined by minimizing the total cost of electricity and chlorine.

3. The radiation from the reflector and lamp is assumed to be perpendicular to the water surface.

Relationship Between Absorption of Secondary Effluent and Chlorine Concentration

In design calculations it is necessary to express the rate equation in terms of concentrations. To do this, A_p must be given in terms of absorption, (A_p°), of secondary effluent before adding chlorine, and in terms of the concentration of free available chlorine.

The absorption of secondary effluent decreases when chlorine is applied, until a large amount of chlorine is added. This is due to formation of low-absorbing substances by reaction between chlorine and pollutants. Such a decolorizing effect more than balances the absorption due to chlorine, until C_{Cl} becomes large. The empirical relationship between the absorption and amount of chlorine added, $(C_{Cl})_t$, was obtained (3) as

$$A_p = \frac{A_p^\circ}{1 + \kappa \left(C_{Cl}\right)_t} \tag{7}$$

where the constant κ depends on the components of secondary effluent (NH_3, --- etc). The relationship between $\left(C_{Cl}\right)_t$ and C_{Cl}, as given in Reference (4), was found to be linear:

$$\left(C_{Cl}\right)_t = a + b\, C_{Cl} \tag{8}$$

where a and b are constants depending on the composition of secondary effluent. From Eqs. (7) and (8), A_p for any secondary effluent may be written as

$$A_p = \frac{A_p^\circ}{a' + b'\, C_{Cl}} \tag{9}$$

where $a' = 1 + \kappa\, a$ and $b' = \kappa\, b$.

The detailed chemical and optical properties of secondary effluent when chlorine is applied are given in (3), (4).

For a trough reactor, with perpendicularly incident light directed inward, the energy absorption per unit length of optical path (of seconda ry effluent) before adding chlorine is obtained from Lambert's law as

$$I(z)A_p^\circ = \frac{\int_o^y (\mu_\lambda I(z) \Sigma \frac{F_\lambda}{F_{tot}} e^{-\mu_\lambda y})dy}{\int_o^y dy}$$

$$= I(z) \frac{1}{y}\left[1 - \sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}} e^{-\mu_\lambda y}\right]$$

where y is the depth of water in the reactor, I(z) is the light intensity at the surface of water, and z is a coordinate along the reactor length. The absorption ratio, A_p°, is given by

$$A_p^\circ = \frac{1}{y}\left[1 - \sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}} e^{-\mu_\lambda y}\right], \text{ cm}^{-1} \qquad (10)$$

Design Equations for Electric Energy Required

Assuming a plug-flow reactor the mass balance of pollutant, for a volume element dV, is

$$\Omega_p \, dV = - q \, d \, (C_p) \qquad (11)$$

where V is the volume of reactor, and q is the volumetric flow rate of effluent in cm^3/sec. If w is the width of the reactor

$$dV = w \, y \, dz \qquad (12)$$

where z is a coordinate along the reactor length. If x_p denotes the conversion of organic carbon,

$$d(C_p) = - C_{p_o} \, d \, x_p \qquad (13)$$

where C_{p_o} is the inlet concentration of pollutant in terms of TOC, g moles/cm^3. The rate of pollutant removal, given in Eqs. (3) and (4), may be written as

$$\Omega_p = \left[\Phi_{p_O} \left(\frac{C_O}{6.5 \times 10^{-8} + C_O} \right) + \Phi_{p_{Cl}} \left(\frac{C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} \right) \right] A_p I(z)/w \qquad (14)$$

where Φ_{p_O} and $\Phi_{p_{Cl}}$ are constants depending on UV spectral distribution, but not on the characteristics of the secondary effluent. Here $I(z)$ is the light intensity at the surface of the trough of water in Einsteins/(cm)(sec). Note that A_p is the average fraction of UV energy absorbed per unit length of optical path. According to condition 2, $A_p y$ is constant with respect to z. Therefore, $\left[1 - \sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}} e^{-\mu_\lambda y} \right]$ is constant.

Then $\mu_\lambda y$ = constant = $\mu_\lambda^o y_o$, and

$$\mu_\lambda y = \alpha_\lambda C_p y = \alpha_\lambda C_{p_o} (1 - x_p) y = \mu_\lambda^o (1 - x_p) y = \mu_\lambda^o y_o \qquad (15)$$

where μ_λ^o is the attentuation coefficient of the feed stream, and y_o is the depth at z = 0. From Eq. (15) we obtain the depth at any conversion as

$$y = \frac{y_o}{1 - x_p} \qquad (16)$$

Substituting Eqs. (9) to (14) in Eq. (11) yields

$$\left[\frac{\Phi_{p_O} C_O}{6.5 \times 10^{-8} + C_O} + \frac{\Phi_{p_{Cl}} C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} \right] \frac{I(z)\, \eta_3\, dz}{a' + b' C_{Cl}} = q\, C_{p_o}\, dx_p \qquad (17)$$

where

$$\eta_3 = A_p^o y = 1 - \sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}} e^{-\mu_\lambda^o y_o} \qquad (18)$$

Note that η_3 represents the fraction of light energy absorbed by secondary effluent. Since $x_p = 0$ at $z = 0$, $x_p = x_{p_f}$ at $z = L$, where L is the length of reactor, integration of Eq. (17) yields

$$\left[\frac{\Phi_{p_O} C_O}{6.5 \times 10^{-8} + C_O} + \frac{\Phi_{p_{Cl}} C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} \right] \frac{\eta_3 I_t}{a' + b' C_{Cl}} = x_{p_f} C_{p_o} q \qquad (19)$$

where

$$I_t = \int_0^L I(z)dz \qquad (20)$$

I_t is the total intensity (Einsteins/sec) at the surface of the water in the trough. We define efficiencies η_1 and η_2 as follows:

$$\eta_1 = \frac{\text{total UV output of lamp (Einsteins/sec)}}{\text{total power input (watts)}} = \frac{I_{ot}}{P_t} \qquad (21)$$

$$\eta_2 = \frac{\text{total UV energy at the surface of the water}}{\text{total UV output of lamp}} = \frac{I_t}{I_{ot}} \qquad (22)$$

Then

$$I_t = \eta_1 \eta_2 P_t \qquad (23)$$

Substituting this expression for I_t in Eq. (19) gives

$$q = \frac{\eta_1 \eta_2 \eta_3 P_t}{x_{P_f} C_{P_o}} \left[\frac{\Phi_{P_0} C_0}{6.5 \times 10^{-8} + C_0} + \frac{\Phi_{P_{Cl}} C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} \right] \frac{1}{a' + b' C_{Cl}}, \text{ cm}^3/\text{sec} \qquad (24)$$

or in terms of gal/hr

$$q' = 0.95\, q \qquad (25)$$

Then the electric power required per gal of secondary effluent will be

$$E = \frac{P_t}{q'} = \frac{1.05\, x_{P_f} C_{P_0}}{\eta_1 \eta_2 \eta_3} (a' + b' C_{Cl}) \Bigg/ \left[\frac{\Phi_{P_0} C_0}{6.5 \times 10^{-8} + C_0} + \frac{\Phi_{P_{Cl}} C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} \right],$$

watts/gal (26)

Equation (26) shows that the electric power requirement decreases with increasing values of the efficiencies η_1, η_2, and η_3. The value of η_2 can be calculated from known reactor dimensions, using the equations in (5). It can also be seen in Eq. (26) that there is an optimum chlorine concentration which gives the minimum required electric energy. This optimum problem will be considered in a later section. The electric power requirement also depends on the spectral distribution of the lamp, since Φ_{P_o} and $\Phi_{P_{Cl}}$ are dependent on the lamp. The higher the conversion,

the higher the feed concentration of pollutants, and the higher the initial chlorine demand, the higher will be the required electric power. Note that Eq. (26) can be applied to all lamps, all secondary effluents*, and at all levels of conversion.

Chlorine Consumption

The concentration of chlorine is maintained constant throughout the reactor. The mass balance of chlorine for a volume element dV is

$$\Omega_{Cl}\, dV = d(F_{Cl}) \tag{27}$$

where $d(F_{Cl})$ is amount of chlorine consumed in dV due to the UV energy absorbed. The rate of free available chlorine disappearance, given in Eqs. (5) and (6), may be written as

$$\Omega_{Cl} = \left[\Phi_{Cl}\, A_{Cl} + \frac{\Phi_{Cl_p}\, C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}} A_p \right] I(z)/w \tag{28}$$

where

$$A_{Cl} = \frac{1}{y}\left[1 - \sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}}\, e^{-\mu_{\lambda Cl} y} \right] \tag{29}$$

It has been shown in Reference (3) that the light energy absorbed by chlorine is very small and independent of pollutant. Then expanding the exponential term in Eq. (29) in a series, and neglecting terms higher than first-order, we obtain

$$A_{Cl} = \sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}}\, \mu_{\lambda Cl} = \bar{\alpha}_{Cl}\, C_{Cl} \tag{30}$$

where $\alpha_{Cl}\left(= \sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}}\, \alpha_{\lambda Cl} \right)$ is the average absorptivity of chlorine. Substituting Eqs. (9), (12), (16), (28), and (30) in Eq. (27) gives

$$d(F_{Cl}) = \left[(\bar{\alpha}_{Cl}\, \Phi_{Cl}) \left(\frac{y_o}{1-x_p} \right) + \frac{\Phi_{Cl_p}\, \eta_3}{(a' + b'\, C_{Cl})(1.1 \times 10^{-9} + C_{Cl})} \right] C_{Cl} I(z) dz \tag{31}$$

*Of the types for which rate measurements were made

Solving Eq. (17) for I(z)dz and substituting this in Eq. (31) gives

$$d(F_{Cl}) = \frac{q\, C_{p_o}\, C_{Cl}\, dx_p}{\left[\dfrac{\Phi_{p_o}\, C_O}{6.5 \times 10^{-8} + C_O} + \dfrac{\Phi_{p_{Cl}}\, C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}}\right]} \left\{ \frac{\bar{\alpha}_{Cl}\, \Phi_{Cl}\, y_o}{\eta_3 (1 - x_p)} \cdot \right.$$

$$\left. (a' + b'\, C_{Cl}) + \frac{\Phi_{Cl_p}}{1.1 \times 10^{-9} + C_{Cl}} \right\} \qquad (32)$$

Integrating Eq. (32) from $x_p = 0$ to $x_p = x_{p_f}$ gives total chlorine consumed due to the light-energy absorbed:

$$F_{Cl} = \frac{q\, C_{p_o}\, C_{Cl}}{\left[\dfrac{\Phi_{p_O}\, C_O}{6.5 \times 10^{-8} + C_O} + \dfrac{\Phi_{p_{Cl}}\, C_{Cl}}{1.1 \times 10^{-9} + C_{Cl}}\right]} \left\{ \frac{\bar{\alpha}_{Cl}\, \Phi_{Cl}\, y_o}{\eta_3} \right. (a' + b'\, C_{Cl}) \left[\ln \right.$$

$$\left. \left. \frac{1}{1 - x_{p_f}} \right] + \frac{x_{p_f}\, \Phi_{Cl_p}}{1.1 \times 10^{-9} + C_{Cl}} \right\}$$

g moles/sec (33)

Or in terms of lbs/hr

$$F'_{Cl} = 564\, F_{Cl}\ , \text{ lbs/hr} \qquad (34)$$

Since the initial amount of chlorine required to obtain C_{Cl} is given by

$$(F_{Cl})_i = (a + b\, C_{Cl})q\ , \text{ g moles/sec}$$

the total amount of chlorine required will be

$$(F'_{Cl})_t = F'_{Cl} + 594\ (a + b\ C_{Cl})\ q'\ ,\ \text{lbs/hr} \qquad (35)$$

Note that Eq. (35) is not restricted to any particular lamp or secondary effluent.

Selection of Lamp

In order to determine what radiant energy source is best suited for secondary-effluent treatment, it is necessary to determine which lamp produces the greatest amount of energy, of the most useful wave length, for the least cost. From Eqs. (26) and (33) we see that lamps which have higher values of Φ_p, η_1, and η_3 give lower operating costs. The efficiency η_1 depends only upon the lamp. On the other hand, the efficiency η_3 depends on both the spectral energy distribution of the lamp and the attenuation coefficients of secondary effluent. Note that the efficiency η_3 can be controlled by the depth of the reactor. High- and low-pressure mercury arcs are normally considered to be the most efficient and most useful sources of ultraviolet energy which are commercially available.

High-pressure lamps emit much more intense radiation than low-pressure lamps of about the same physical size. Although the quantum yield is greater for a low-pressure lamp, the number of lamps required would be much greater for the high-pressure variety. However, the useful life of the high-pressure mercury arc is relatively short--usually less than 1,000 hrs. Low-pressure lamps have long lifetimes -- usually 7,500 hrs, or longer. High-pressure lamps are the more expensive but do not have higher efficiencies, η_1. Further, high-pressure lamps require air cooling to obtain the best performance.

Since electricity is a major operating expense, we should compare the electric power requirement for both lamps. Neglecting the contribution of oxygen to the rate, the ratio of the electric power requirements for both lamps [as obtained from Eq. (26)] and for the same absorption will be

$$\frac{(E)_{Hp}}{(E)_{Lp}} = \left[\frac{(\eta_1)_{Lp}}{(\eta_1)_{Hp}}\right]\left[\frac{\Phi_{p_{Cl}\ Lp}}{\Phi_{p_{Cl}\ Hp}}\right] \qquad (36)$$

The high- and low-pressure lamps which have the highest efficiencies η_1 among the lamps available are, respectively,

(a) 2100-watt, high-pressure, mercury lamp (78A Hanovia):
$\eta_1 = 1.054 \times 10^{-6}$ Einsteins/(watt)(sec)

and

(b) 65-watt, low-pressure mercury lamp (G64T6 General Electric)

$\eta_1 = 5.86 \times 10^{-7}$ Einsteins/(watt)(sec)

The values for $\Phi_{p_{Cl}}$ are given in Eqs. (3) and (4). Substituting numerical values for η_1 and $\Phi_{p_{Cl}}$ in Eq. (36), we see that the electric power requirement is three times higher for the high-pressure lamp. Further, the ratio of chlorine consumption for both lamps [obtained from Eq. (33)] and for the same absorption is approximately given by

$$\frac{\left(F_{Cl}\right)_{Hp}}{\left(F_{Cl}\right)_{Lp}} = \frac{\left(\Phi_{p_{Cl}}\right)_{Lp}}{\left(\Phi_{p_{Cl}}\right)_{Hp}} \left[\frac{\left(\Phi_{Cl}\right)_{Hp}}{\left(\Phi_{Cl}\right)_{Lp}}\right] \qquad (37)$$

The values for $\Phi_{p_{Cl}}$ and Φ_{Cl} are given in Eqs. (3) to (6). Substitution of numerical values for $\Phi_{p_{Cl}}$ and Φ_{Cl} in Eq. (37) shows that chlorine consumption is 3.4 times larger for the high-pressure lamp. Therefore, the best source appears to be a 65-watt, low-pressure, mercury lamp (G64T6 General Electric). This lamp will be chosen for economic calculations.

Design Calculations for Treatment of 1,000 gal/hr Secondary Effluent

This section shows the form of design calculations and does not provide a practical design for a small (1,000 gal/hr) unit. The numerical calculations will be based on the following design conditions:

(a) 1,000 gal/hr of secondary effluent from the Howe Avenue treatment plant in Sacramento, California ($q' = 1{,}000$ gal/hr).

(b) Organic pollutant is to be reduced from 10 mg/liter (TOC) to 4[C_{p_o} = 10 mg/liter, or $\frac{10}{12} \times 10^{-6}$ g moles/cm^3, $x_{p_f} = 0.6$].

(c) The numerical values for a, b, and κ for the secondary effluent are:
$a = 1.38 \times 10^{-7}$, $b = 1.52$, $\kappa = 2.5 \times 10^{6}$
These values were obtained for the secondary effluent from the Howe Avenue plant in Sacramento [see References (3) and (4)].

(d) The reflectance (ρ) of reflector is 0.7, as found by measurement for aluminum [see Reference (5)].

(e) The concentration of oxygen is 0.28×10^{-6} g moles/cm^3, corresponding to saturation of water with air at room temperature.

(f) The costs of electricity and chlorine chosen are 1¢/KWH and 3.75¢/lb, respectively. The numerical values for the parameters in the rate equations for a low-pressure lamp [see Eqs. (4) and (6)] are;

I_{ot}	$= 3.81 \times 10^{-5}$	Einsteins/sec
Φ_{p_0}	$= 2.07 \times 10^{-2}$	g moles/Einstein
$\Phi_{p_{Cl}}$	$= 0.101$	g moles/Einstein
Φ_{Cl}	$= 0.52$	g moles/Einstein
Φ_{Cl_p}	$= 0.057$	g moles/Einstein
$\bar{\alpha}_{Cl}$	$= 2.6 \times 10^{5}$	cm^2/g mole

The efficiency η_2, given in (5), may be written as

$$\eta_2 = \zeta \left\{ \left[\left(\frac{1+\delta}{2}\right)^2 + \left(\frac{\alpha \left(1+\frac{1}{16\gamma^2}\right)}{2} \right)^2 \right]^{1/2} - \left[\left(\frac{1-\delta}{2}\right)^2 + \left(\frac{\alpha \left(1+\frac{1}{16\gamma^2}\right)}{2} \right)^2 \right]^{1/2} \right\}$$

$$+ (1-\zeta) \left\{ \left[\left(\frac{1+\delta}{2}\right)^2 + \left(\frac{\alpha \left(1-\frac{1}{16\gamma^2}\right)}{2} \right)^2 \right]^{1/2} - \left[\left(\frac{1-\delta}{2}\right)^2 + \left(\frac{\alpha \left(1-\frac{1}{16\gamma^2}\right)}{2} \right)^2 \right]^{1/2} \right\} \qquad (38)$$

where

$$\zeta = \frac{1}{2} + \frac{1}{\pi} \tan^{-1} \left[2\gamma \left(1 - \frac{1}{16\gamma^2} \right) \right]$$

$$\delta = \frac{L}{\ell}$$

$$\gamma = \frac{h}{w}$$

$$\alpha = \frac{2h}{\ell}$$

L = length of reflector (= reactor), ft

ℓ = length of lamps, ft

h = height of reflector above water surface, ft

w = width of reflector (= reactor), ft

Figure 2 shows the efficiency η_2 as functions of the ratios, γ and α, when $\rho = 0.7$ and $\delta = 1.3$. We see from this figure that the efficiency, η_2, increases as both the ratio of height to width and the ratio of height to lamp length decreases. This indicates that wide, low, and long reflectors give higher efficiencies. One restriction to height is the necessary distance between the lamp and the surface of water to prevent excessive deposits on the lamp. We choose six inches as the minimum safe distance. Then from the equation for a parabola, the height in terms of γ is

$$h = \frac{0.5}{1 - \frac{1}{16\gamma^2}} \text{ , ft} \qquad (39)$$

Since the power input per unit length of low-pressure lamp is small (0.411 watts/cm), the length of lamp required for 1,000 gal/hr wastewater treatment is expected to be very large. Therefore, $\alpha = \frac{2h}{\ell} \approx 0$ and we choose $\delta = \frac{L}{\ell} = 1$. As noted from Figure 2, the efficiency η_2 increases when the ratio γ decreases. However, for $\gamma < 0.3$ the change in η_2 with respect to γ is very small. Hence $\gamma = 0.3$ is chosen as the optimum value. Note that the volume of the reactor increases as γ decreases. Substituting these values for α, γ, and δ, and $\rho = 0.7$, in Eq. (38) gives

$$\eta_2 = 0.833$$

The low-pressure mercury lamp emits very little radiation other than that at 2537 Å. Hence, this lamp may be considered as a monochromatic ultraviolet source. Then the efficiency, η_3 (Eq. (18)), becomes

$$\eta_3 = 1 - e^{-\mu_\lambda^o y_o}$$

where $\mu^o_{2537} = 0.264$ for the secondary effluent from the Howe Avenue plant (corresponding to TPC of 10 mg/liter.

With the chosen design conditions, substituting values for η_1, η_2, η_3 and the parameters of the rate equations in Eqs. (26) and (35) gives the electric power and the amount of chlorine required. The results for a treatment rate of 1,000 gal/hr, as functions of the chlorine concentration and the initial depth of reactor, are:

$$E = \frac{107.55\ (1.345 + 0.038\ C_{Cl} \times 10^8)(0.11 + C_{Cl} \times 10^8)}{(1 - e^{-0.264\ y_o})(0.185 + 11.78\ C_{Cl} \times 10^8)} \text{ , KWH} \qquad (40)$$

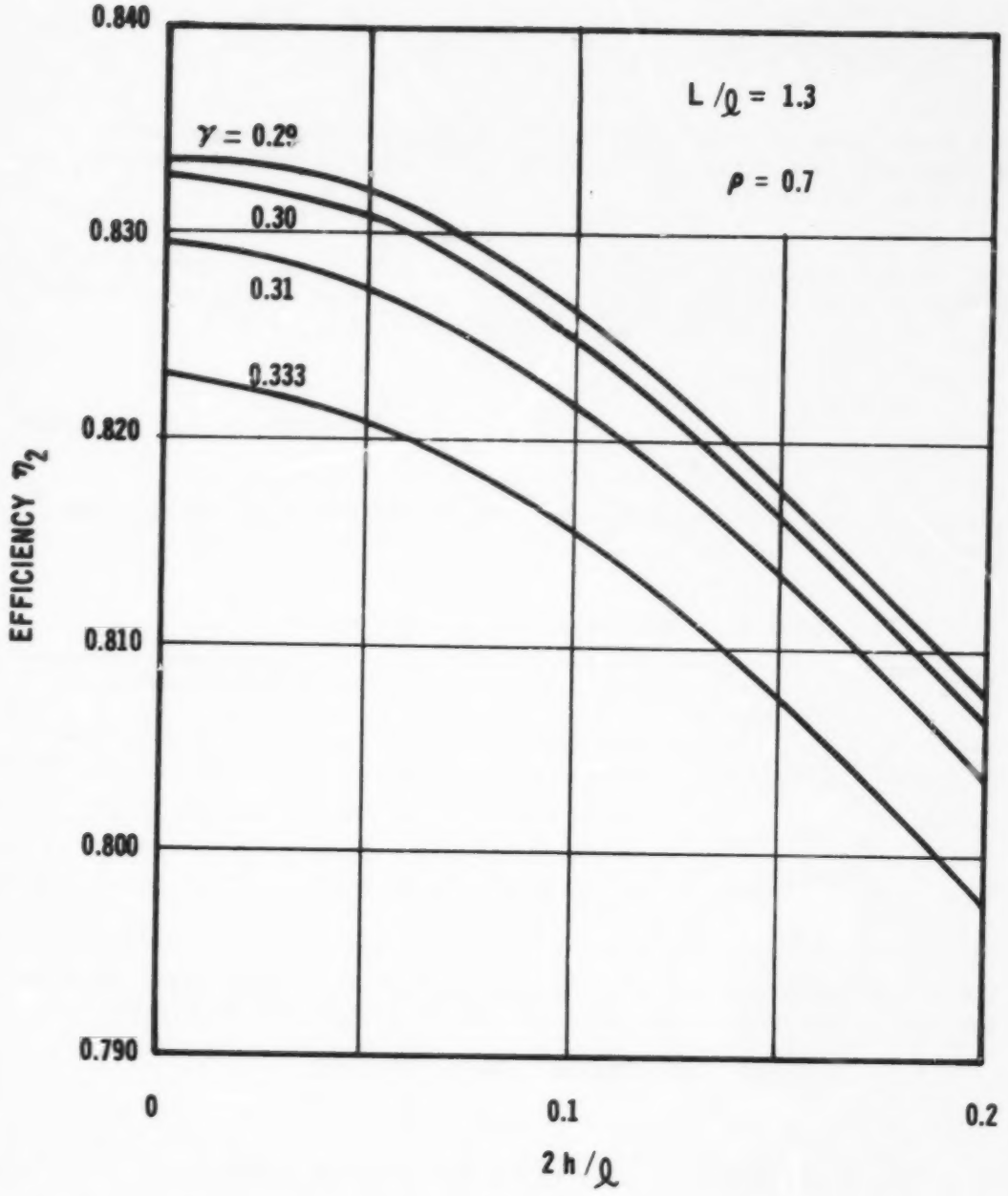


Figure 2. Effect of Ratio of Height to Width on Efficiency η_2

$$(F'_{Cl})_t = \frac{0.061\, y_o\, C_{Cl} \times 10^8\, (1.345 + 0.038\, C_{Cl} \times 10^8)(0.11 + C_{Cl} \times 10^8)}{(1 - e^{0.264\, y_o})(0.185 + 11.78\, C_{Cl} \times 10^8)}$$

$$+ \frac{1.69\, C_{Cl} \times 10^8}{0.185 + 11.78\, C_{Cl} \times 10^8} + 0.009\, C_{Cl} \times 10^8 + 0.082,\ \text{lbs/hr} \qquad (41)$$

Since the costs of electricity and chlorine are 1¢/KWH and 3.75¢/lb, respectively, the total cost of electricity and chlorine is obtained from Eqs. (40) and (41) as

$$S_{¢} = \frac{(1.345 + 0.038\, C_{Cl} \times 10^8)(0.11 + C_{Cl} \times 10^8)(107.55 + 0.0229\, y_o\, C_{Cl} \times 10^8)}{(1 - e^{0.264\, y_o})(0.185 + 11.78\, C_{Cl} \times 10^8)}$$

$$+ \frac{6.348\, C_{Cl} \times 10^8}{0.185 + 11.78\, C_{Cl} \times 10^8} + 0.0338\, C_{Cl} \times 10^8 + 0.307,\ \text{Cents} \qquad (42)$$

The total costs calculated from Eq. (42) at various C_{Cl} and y_o are summarized in Table 1

Table 1. Electricity and Chlorine Costs

	Depth y_o, cm	Cost, cents/1,000 gal						
		7	9	11	13	15	17	19
$C_{Cl} = 0.5 \times 10^{-8}$, g moles/cm^3		18.46	17.23	16.60	16.27	16.09	16.00	16.02
$= 1.0 \times 10^{-8}$		17.49	16.37	15.80	15.51	15.38	15.30	15.34
$= 1.5 \times 10^{-8}$		17.38	16.31	15.78	15.51	15.40	15.39	15.44
$= 2.0 \times 10^{-8}$		17.50	16.45	15.94	15.71	15.63	15.63	15.72

The optimum chlorine concentration and optimum depth which give the minimum cost of electricity and chlorine are found from Eq. (42) to be

$C_{Cl} = 1.1 \times 10^{-8}$ g moles/cm^3 (0.78 mg/liter)

$y_o = 17$ cm (0.56 ft)($\eta_3 = 0.9887$)

Cost = 15.21¢ (Electricity 13.88¢ and chlorine 1.33¢)

With these values, the electric power required for 1,000 gal/hr wastewater treatment is E = 13.88 KWH/1,000 gal. If four parallel lamps are placed at the focus of the parabolic reflector, the power input per foot

(using 65-watt low-pressure lamps) is 53.8 watts/ft. Then, the total length of lamps needed is

$$\ell = \frac{13.88 \times 10^3}{53.8} = 257 \text{ ft}$$

The amount of chlorine required can be found from Eq. (41):

$$(F'_{Cl})_t = 0.355 \text{ (lbs/hr)}$$

The optimum values for geometry and chlorine concentration for 1,000 gal/hr of secondary effluent from the Howe Avenue plant are summarized as follows:

(1) Reflector (parabola)
width - 4.92 ft
height - 1.64 ft
distance between lamp and surface of water = 0.5 ft
length = 257 ft
$\eta_2 = 0.833$

(2) Lamp: 65 watt, low-pressure mercury lamp (G64T6 General Electric)
total length = 257 ft
total number = 214

(3) Reactor (Trough)
depth: inlet = 0.56 ft
outlet = 1.4 ft
length = 257 ft (length parallel to lamps)
width = 4.92 ft
volume = 1240 ft^3
concentration of chlorine = 0.78 mg/liter

(4) Operating cost (electricity and chlorine)
electricity = 13.88 KWH or 13.88¢
chlorine = 0.355 lbs/hr or 1.33¢
Total = 15.21¢

From Eq. (26) the electric power requirement is seen to depend upon the absorption characteristics of secondary effluent (with chlorine added) and upon the initial chlorine demand of the water. For example, if the initial chlorine demand is negligible, i.e. a = 0, then the electric energy requirement for 1,000 gal/hr of water treated will be reduced from 13.88 KWH to 10.3 KWH. Hence, a photochemical treatment process would be more suitable for low, initial-chlorine-demand water, and for secondary effluent for which the absorption does not change much when chlorine is added (i.e. small value of κ).

Equation (26) also shows that a large reduction in electric power requirement would be possible by improving the UV output of the lamp. For example, if η_1 for the low-pressure lamp could be doubled, i.e.

increased from 5.86×10^{-7} Einsteins/(watt)(sec) to 1.172×10^{-6} Einsteins/(watt)(sec), the electric power requirement for 1,000 gal/hr treatment would be reduced to 6.94 KWH.

The Hanovia Lamp Company attempted to build a more intense low-pressure lamp with higher η_1 in response to our request, but the first attempt did not succeed.

Design Calculations for One Million gal/day Treatment Plant

The plant is designed to reduce TOC from 10 to 4 mg/liter, using G64T6, low-pressure, mercury-arc lamps. This lamp is in commercial production and has the highest efficiency, η_1, among low-pressure lamps available. The UV output is 18.5 watts for an electrical power input of 65 watts. A plant would handle one million gal/day. All other design conditions are the same as for the 1,000 gal/hr calculations given in the previous section. The concentration of chlorine is maintained constant at C_{Cl} = 0.78 mg/liter throughout reactor.

(1) Determination of electric energy and chlorine requirements

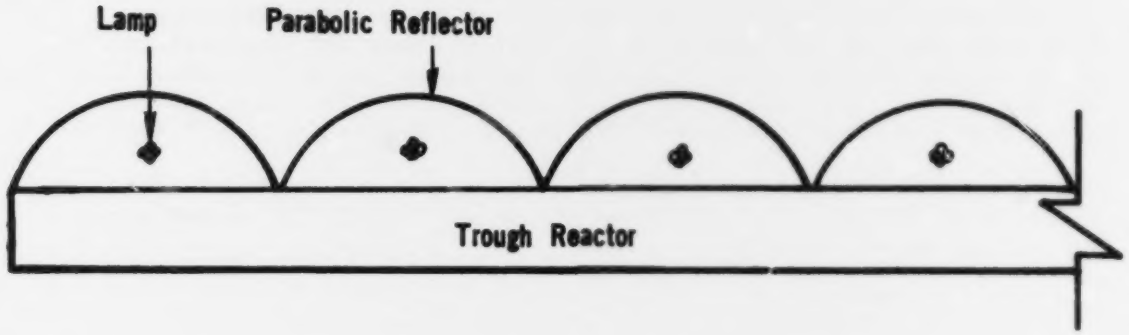
The efficiency, η_2, is the only parameter in the design equation which may be affected by scale-up. However, for low-pressure lamps the total length of lamps in one reflector is long enough for the ratio $\alpha = \frac{2h}{\ell}$ to be negligible. Note that η_2 does not change for $\alpha < 0.05$, even if the total length of lamps is changed, provided that other parameters in Eq. (38) are constant. In order that η_2 be high, a reflector is chosen which has the same height and width as for the 1,000 gal/hr design calculations. Hence, a value of $\eta_2 = 0.833$ will be used. With these conditions the electric energy and chlorine requirements are directly proportional to the flow rate. For a feed rate of one-million gal/day, we obtain, from Eqs. (26) and (35),

Electricity = 578 KWH
Chlorine = 1.48 lbs/hr

(2) Reactor-reflector geometry and plant description

The reactor shown in Figure 3 represents an arrangement such that $\eta_2 = 0.833$, which is the highest efficiency obtainable for the given design conditions. Then the total number of lamps is 8,890. The ratio of width to length of the reactor is unimportant as long as $\alpha < 0.05$ and the total number of lamps = 8,890. As a practical possibility we choose

width of reactor = 172 ft
length of reactor = 309 ft



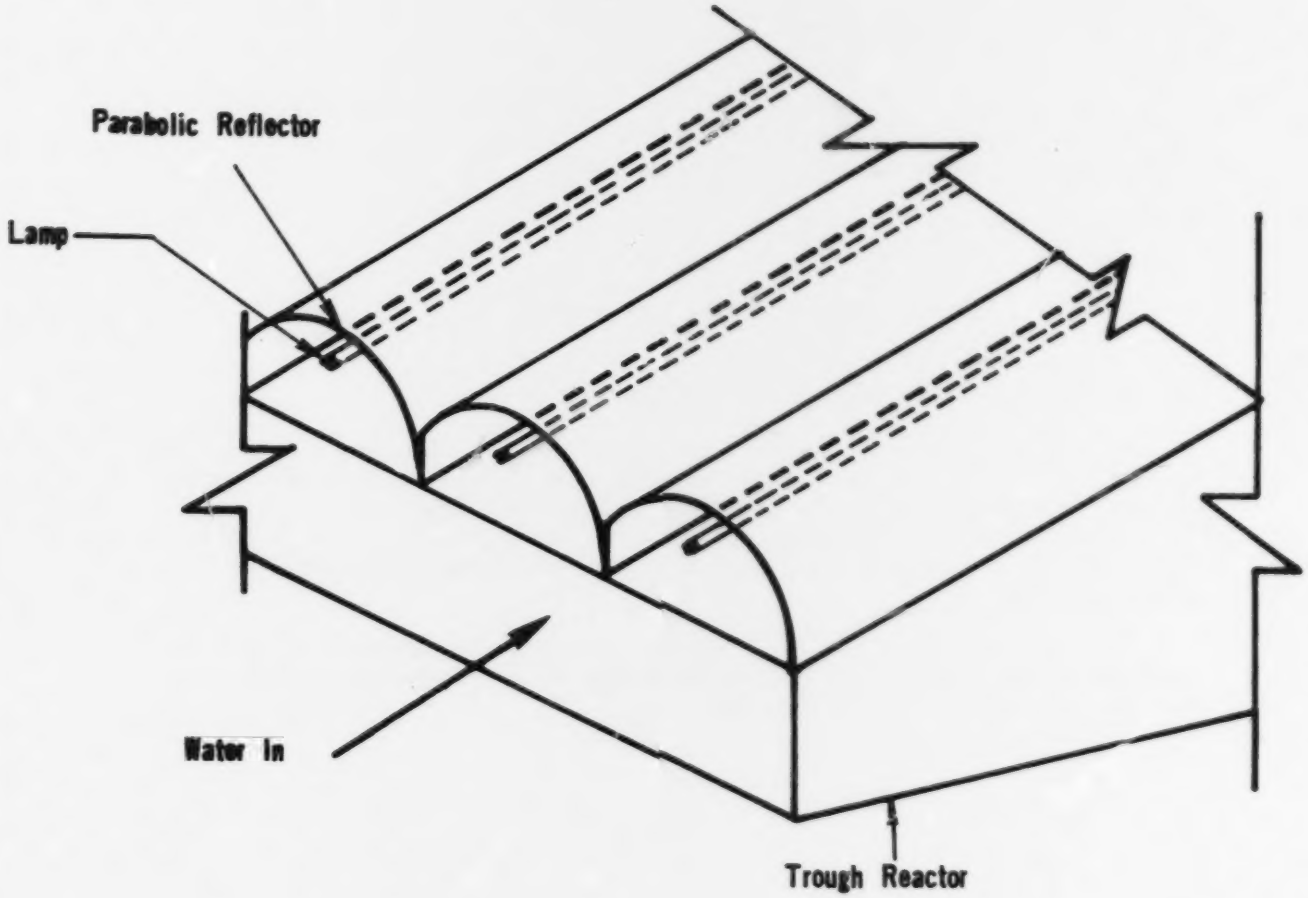


Figure 3. Arrangement of Reactor and Reflectors.

As shown in Figure 3, four parallel lamps are placed near the focus of the reflector. The reactor cover consists of 35 parabolic reflectors. Each reflector has 254 lamps and then about 64 lamps are connected in series at the line of focus. The inlet and outlet depths of reactor are chosen to be the same as for 1,000 gal/hr treatment, since the concentration of free available chlorine is maintained at C_{Cl} = 0.78 mg/liter.

The optimum values for reactor-reflector geometry (to reduce the TOC from 10 to 4 mg/liter) for secondary effluent from Howe Avenue plant are summarized as follows:

(1) Reflector (parabolic)
- width = 4.92 ft
- height = 1.64 ft
- distance between lamp and surface of water = 0.5 ft
- length = 309 ft
- number of reflectors = 35
- η_2 = 0.833

(2) Lamp: 65 watt, low-pressure mercury lamp (G64T6 General Electric)
- total number of lamps required = 8,890
- number of lamps in each reflector = 254

(3) Reactor (trough)
- depth: inlet = 0.56 ft
  outlet = 1.4 ft
- length = 309 ft (length parallel to lamps)
- width - 172 ft
- volume = 52,400 ft^3
- concentration of chlorine = 0.78 mg/liter

A process flowsheet is given in Figure 4. Chlorine would be supplied as vapor from a 1,400 gal liquid chlorine tank. Automatic, flow-control equipment would regulate the chlorine flow to each injection point in order to maintain the constant chlorine concentration (C_{Cl} = 0.78 mg/liter). There are 35 and 64 injection points across and along the trough reactor, respectively.

Air would be supplied (from an air compressor) uniformly to each injection point. The number and arrangements of air injection points are the same as those for chlorine.

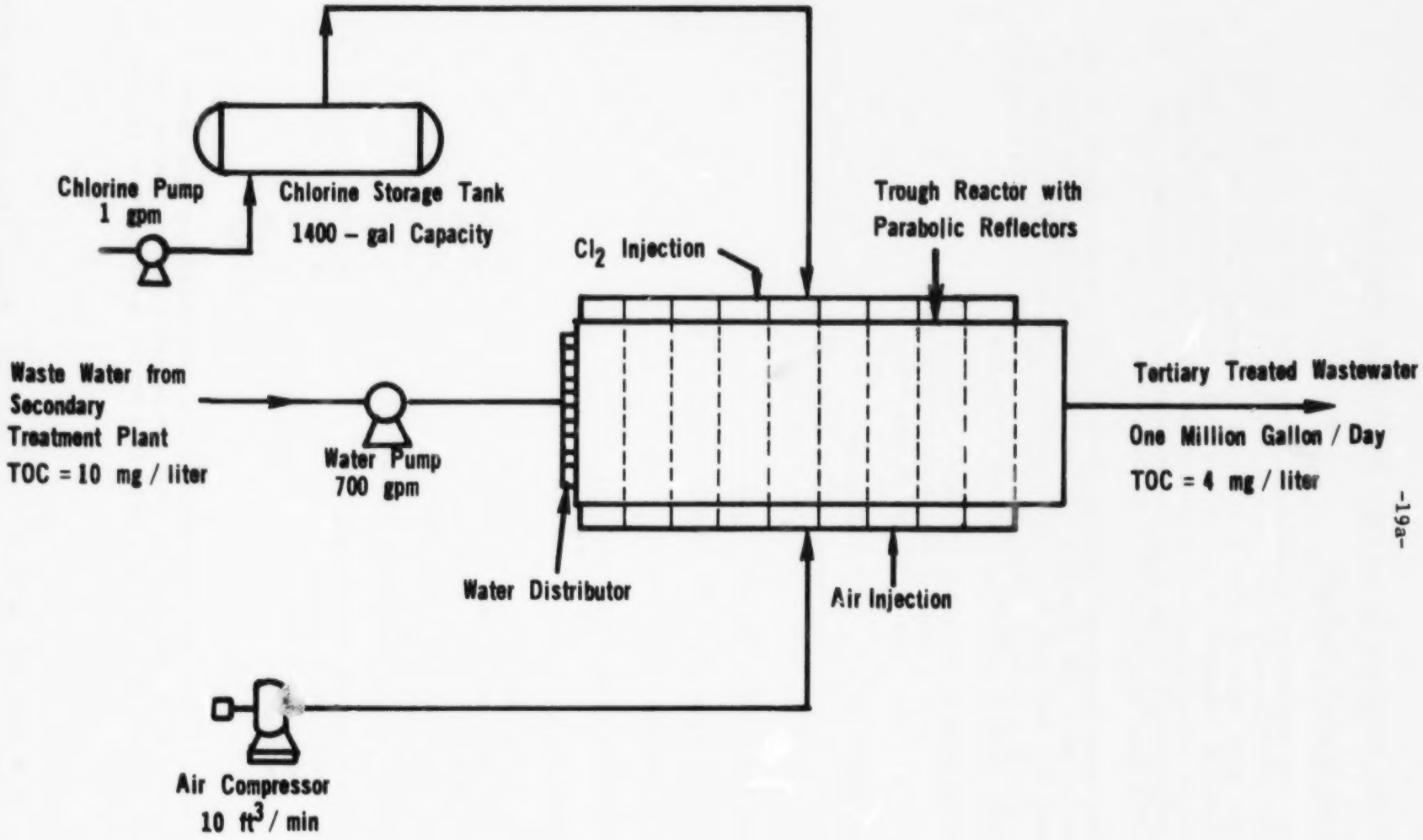


Figure 4. Process Flowsheet for Photochemical Treatment of Secondary Effluent (One Million gal / Day)

V. EVALUATION OF ECONOMICS FOR TERTIARY TREATMENT

A. Fixed-Capital Investment

Because prices may change considerably with time and location, a detailed calculation of costs is not warranted. The results presented here are estimates, but they do indicate the general level of costs for photochemical treatment at its present stage of development. A reliable estimate of costs would require engineering drawings, specifications, and site surveys would be required for construction of a treatment plant. The chief reference for costs used here was Peters and Timmerhaus (6).

The plant investment has been estimated for a plant capacity of one-million gal/day. The design conditions and detailed dimensions of major equipment are described in the previous part (process design) of the report. Figure 4 shows the major equipment. The cost of installed major equipment items is $185,400, itemized as follows:

1. Trough reactor; 6" thick concrete
   total surface area = 53,040 ft^2
   cost = $88,500 ($15/sq yd)

2. Reflectors (35); $\frac{1}{16}$" thick aluminum
   total surface area = 150,000 ft^2
   cost = $82,500 ($0.55/ft^2)

3. Wastewater pump; 700 gpm, $2,000

4. Chlorine pump (stainless steel); 1 gpm, $1,800

5. Air compressor; 10 ft^3/min; $5,600

6. Chlorine storage tank; 1,400 gal, $5,000

Other plant costs are:

1.	Instrumentation and controls	$ 33,300
2.	Piping (installed)	38,900
3.	Electrical (installed)	27,800
4.	Buildings (including service)	44,500
5.	Yard improvements	11,000
6.	Service facilities (installed)	55,600
7.	Land	5,500
8.	Engineering and supervision	50,000
9.	Construction expense	56,000
10.	Contractor's fee	11,100
11.	Contingency	44,100
		$377,800

This gives a total plant investment of $563,200, excluding lamp costs. Since a life expenctancy of the G64T6 low pressure mercury lamp is one year, it is best to treat lamp costs as a yearly operating cost (in the next section).

B. Annual Operating Expenses

These expenses include direct treatment cost and fixed charges. Annual costs are based on 360 working days.

The treatment cost was estimated as follows:

	Cost per year	¢/(1,000 gal)
Operating labor: 2 men/shift	$ 64,000	17.76
Raw material		
Chlorine: 355 lbs/day at 3.75¢/lb	4,800	1.33
Utilities		
Electricity for UV energy: 578 KWH at 1¢/KWH	50,000	13.88
Electricity for pumps, compressor & buildings	6,000	1.66
Mercury lamps: 8,890 at $14*	124,000	34.20
Maintenance and repairs, at 3% of fixed capital investment	16,700	4.64
Operating supplies	2,500	0.69
	$268,000	74.16¢

Fixed charges on the investment of $563,200 have been estimated as

	Cost per year	¢/(1,000 gal)
Depreciation over 20 years	$ 27,800	7.73
Insurance at 1%	5,600	1.55
Interest at 4%	22,200	6.10
	$ 55,600	15.38¢
Total	$323,600	89.5

The total treatment costs are $323,600 annually, including fixed charges on investment and operating costs. The unit-cost total is 89.5¢/(1,000 gal). Note that the major treatment costs are: lamp, 34.20¢/(1,000 gal), electricity, 13.88¢/(1,000 gal), and labor, 17.76¢/(1,000 gal). Lamp and electricity costs depend mainly on the electric power requirement. MRI (1) has estimated these costs to be much lower: lamp, 1.7¢/(1,000 gal); electricity, 1.06¢/(1,000 gal); labor, 1.86¢/(1,000 gal). The chief reason for this discrepancy is the difference in quantum yield. The quantum yield used by MRI was 1.3 g moles/Einstein. On the other hand, the maximum quantum yield found in this work is 0.12 g moles (TOC)/Einstein (see Eq. (4)). As a result the total number of lamps and the electricity required for 1,000 gal are each one order of magnitude higher than the estimates by MRI.

Substantial reductions in cost would be possible if more intense, longer-life, and higher UV-output (per unit power input) low-pressure mercury lamps were available. Among these possibilities, higher UV-output per unit power input (η_1) is most valuable in reducing the treatment costs. The labor cost per 1,000 gal could be reduced for higher water throughputs.

*Single lamp cost at present is $32.00. It is estimated that large purchases could reduce this to $14/lamp.

VI. ACKNOWLEDGEMENT

Dr. C. P. Jeffreson, Dr. V. Schorr, Dr. B. Boval and Dr. V. Hancil assisted importantly in carrying out the experimental measurements and analysis of results.

The support of the project by the Environmental Protection Agency, and the help provided by Mr. Robert H. Wise, the Grant Project Officer, is also acknowledged.

VII. NOMENCLATURE

A	absorption ratio, cm^{-1} A_p = absorption ratio for secondary effluent A^o_p = absorption ratio for the feed stream of secondary effluent before chlorine is added A_{Cl} = absorption ratio for chlorine
a	initial chlorine-demand parameter in Eq. (8), g moles/cm^3 $a' = 1 + \kappa a$
b	constant in Eq. (8) $b' = \kappa b$
C	concentration, g moles/cm^3 C_p = concentration of pollutant C_{Cl} = concentration of free available chlorine C_O = concentration of oxygen $(C_{Cl})_t$ = amount of chlorine added initially
E	electrical energy requirement, watts/gal
F_λ	fraction of UV energy emitted by lamp of wave length λ
F_{tot}	total UV energy emitted by lamp
F_{Cl}	amount of chlorine consumed due to UV energy, g moles/sec; F'_{Cl}, lbs/hr $(F'_{Cl})_t$ = total amount of chlorine required, lbs/hr $(F_{Cl})_i$ = amount of chlorine required to obtain given C_{Cl}, lbs/hr
h	height of reflector above water, ft
I(z)	intensity of radiation, at the surface of water at z, Einstein/(cm)(sec)
I_t	total intensity at the surface, Einsteins/sec
I_{ot}	total UV output from lamp, Einsteins/sec
I_{tot}	total intensity of UV energy at the reactor wall, Einsteins/(cm^2)(sec)
L	length of reactor (or reflector), ft
ℓ	total length of lamps in one reflector, ft
P_t	total electric power input, watts

q volume flow rate of secondary effluent, cm^3/sec
q' = gal/hr

x conversion; x_p refers to organic carbon pollutants

v volume of reactor, cm^3

w width of reflector (or reactor), cm

y depth of reactor, cm

z axial distance, cm

TOC concentration of total organic carbon, g moles/cm^3, or mg/liter

Greek

α ratio of height of reflector to one half of lamp length $h/(\ell/2)$

α_λ absorptivity at wave length λ, cm^2/g mole

$\bar{\alpha}$ = average absorptivity = $\sum_{\lambda_1}^{\lambda_2} \frac{F_\lambda}{F_{tot}} \alpha_\lambda$

δ ratio of length of reflector to that of lamp = L/ℓ

γ ratio of height to width of reflector = h/w

Ω rate of reaction, g moles/(cm^3)(sec)
Ω_{Cl} = rate of free available chlorine disappearance
Ω_p = rate of organic pollutant removal

λ wave length, cm

Φ quantum yield, g moles/Einstein
Φ_{p_O} = quantum yield for contribution of oxygen to rate of pollutant disappearance
$\Phi_{p_{Cl}}$ = quantum yield for contribution of free available chlorine to rate of pollutant disappearance
Φ_{Cl_p} = quantum yield for contribution of pollutant to rate of free available chlorine disappearance
Φ_{Cl} = quantum yield for free available chlorine disappearance due to A_{Cl}

μ_λ attenuation coefficient at wave length λ, cm^{-1}
μ_λ^o = feed stream of secondary effluent
$\mu_{\lambda Cl}$ = chlorine

η_1 fraction of the electrical energy input which is emitted in the UV wave-length region of the lamp

η_2 fraction of UV energy leaving lamp which strikes the surface of the secondary effluent

η_3 fraction of the incident UV energy which is absorbed by secondary effluent

ρ reflectance

γ parameter in Eq. (38)

Subscripts

Cl free available chlorine

f final, or exit conditions

Hp high-pressure mercury lamp

Lp low-pressure mercury lamp

o feed or entrance conditions

O oxygen

p pollutant

t total

Superscripts

o feed condition before chlorine is added

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